LA-UR-04-6421

Approved for public release; distribution is unlimited.

Title:

TARGETRY AT THE LANL 100 MeV ISOTOPE PRODUCTION FACILITY: LESSONS LEARNED FROM FACILITY COMMISSIONING

Author(s):

F. M. Nortier, M. DeJohn, M. E. Fassbender, V. T. Hamilton, R. C. Heaton, D. J. Jamriska, J. J. Kitten, J.W. Lenz, C. E. Lowe, C.F. Moddrell, L. M. McCurdy, E. J. Peterson, L. R. Pitt, D. R. Phillips, L. L. Salazar, P.A. Smith and F. O. Valdez

Submitted to:

Americas Nuclear Energy Symposium 2004, October 3-6, 2004, Miami Beach, Florida



_



Los Alamos National Laboratory, an affirmative action/equal opportunity employer, is operated by the University of California for the U.S. Department of Energy under contract W-7405-ENG-36. By acceptance of this article, the publisher recognizes that the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or to allow others to do so, for U.S. Government purposes. Los Alamos National Laboratory requests that the publisher identify this article as work performed under the auspices of the U.S. Department of Energy. Los Alamos National Laboratory strongly supports academic freedom and a researcher's right to publish; as an institution, however, the Laboratory does not endorse the viewpoint of a publication or guarantee its technical correctness.



TARGETRY AT THE LANL 100 MeV ISOTOPE PRODUCTION FACILITY: LESSONS LEARNED FROM FACILITY COMMISSIONING *

F.M. Nortier¹, M. E. Fassbender⁺¹, M. DeJohn³, V. T. Hamilton¹, R. C. Heaton¹, D. J. Jamriska¹, J. J. Kitten¹, J.W. Lenz², C. E. Lowe¹, C.F. Moddrell³, L. M. McCurdy¹, E. J. Peterson¹, L. R. Pitt¹, D. R. Phillips¹, L. L. Salazar¹, P.A. Smith³ and F. O. Valdez¹

¹Los Alamos National Laboratory, MS J514, Los Alamos, New Mexico 87545, USA.

²John W. Lenz & Associates, 412 Muskingum Road, Waxahachie, TX 75165

³P.A. Smith Concepts & Designs,

1475 Central Ave. Suite 250,Los Alamos, New Mexico 87544

Abstract

The new Isotope Production Facility (IPF) at Los Alamos National Laboratory has been commissioned during the spring of 2004. Commissioning activities focused on the establishment of a radionuclide database, the review and approval of two specific target stack designs, and four trial runs with subsequent chemical processing and data analyses. This paper highlights some aspects of the facility and the targetry of the two approved target stacks used during the commissioning process.

[`]LA-UR#

corresponding author, e-mail: mifa@lanl.gov

Introduction, Materials and Methods

On December 23, 2003, the first 100 MeV proton beam was delivered to the new irradiation facility for the production of radioisotopes at Los Alamos National Laboratory. For the next four months, activities at the new Isotope Production Facility (IPF) [1] focused on commissioning. Pursuant to the demonstration of safe, compliant and reliable operation, particular attention was given to the operation of the beam line, the target station and the targetry [2] at the maximum design parameters.

Two target stack designs were approved for receiving beam during the facility's commissioning process: the first stack, referred to as "Dummy" target stack, consisted of three durable metal targets with niobium in the high energy slot, zinc in the medium energy, and aluminum in the low energy slot. Each target was capable of accepting 250 μ A of beam [2,3]. This stack was used to demonstrate safe and reliable operation at 250 μ A, the maximum design average beam current. The second target stack, the "Prototype" target stack, consisted of target disks intended for the production of bulk radionuclide quantities: two stainless-steel encapsulated rubidium chloride (RbCl) targets for production of ⁸²Sr occupied the high energy and the medium energy slots, while a niobium encapsulated gallium metal target for production of ⁶⁸Ge occupied the low energy slot.

Thermal conductivity analyses were performed using computational fluid dynamics (CFD). Basic assumptions for the CFD calculations included a typical ring shaped beam profile as produced by sweeping the Gaussian proton beam in a circle across the target face. Other assumptions were a 250 μ A average beam current, 625 μ s pulse length at 30 Hz and a 30 GPM bulk cooling water flow rate [3]. One stack of the first type and three of the second type were successfully irradiated during commissioning. During and after bombardments, individual targets were visually inspected for signs of thermal deterioration. These targets were disassembled and transported to the Hot Cell facility for chemical processing.

Figures 1 and 2 depict a schematic view of the IPF construction. The target irradiation chamber is located in the basement of the facility. A retrieval system allows the remote controlled removal of the irradiated item through a well: The target carrier is hauled into a preparation hot cell, where it can be safely handled and packaged for transportation.

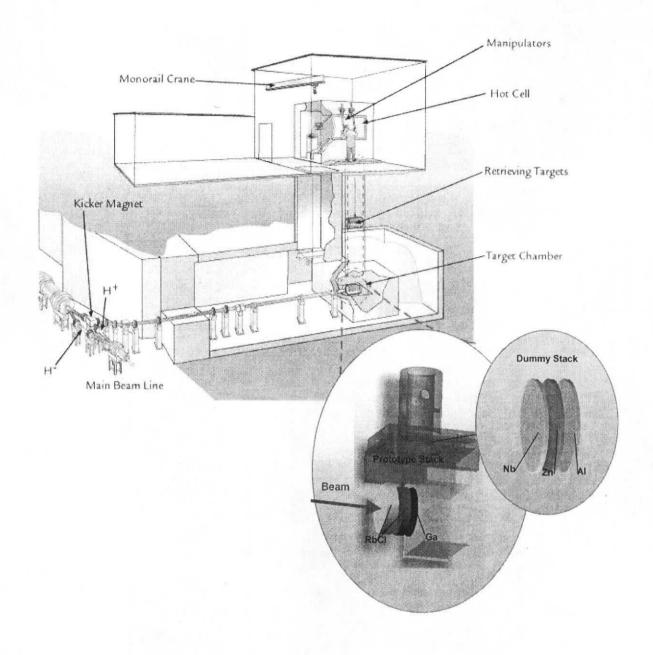


Fig 1. Schematic view of the Isotope Production Facility (IPF) at Los Alamos; the enlarged view shows the two ("Dummy" and "Prototype") target stack designs approved for the commissioning of IPF

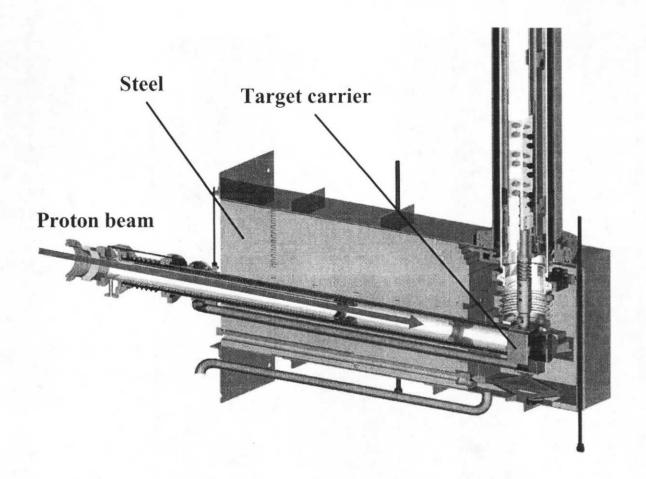


Fig 2. Schematic view of IPF's proton bombardment part: the target carrier is located in a cube-shaped steel box, which, in turn, is surrounded by concrete

Results and Discussion

Table 1 shows the summarized beam parameters for the targets used in this work. Energy values were originally calculated assuming the density of solid RbCl salt. Thermal analysis however, revealed that the salt melts during irradiation. Thus, recalculations assuming the density of liquid RbCl were performed resulting in different beam entrance and exit energies.

Table 2 presents the projected maximum temperatures reached in the two CFD modeled targets of the "Prototype stack". Rubidium chloride was only modeled for the high energy slot. In this case, the maximum temperature exceeds the material boiling point, which is reached at a beam current of $130 \mu A$. Consequently, such targets should

be irradiated at beam currents not exceeding $100\mu A$ in order to avoid capsule rupture due to pressure build-up.

Table 1. CFD modeled maximum temperatures reached in target materials of the "Prototype" target

| Material | Energy slot | Beam current | Max. temp. | m.p. | b.p. |
|----------|-------------------|-----------------|------------|------|------|
| | [MeV] | [μΑ] | [oC] | [oC] | [oC] |
| RbCl | $[92.4 \to 70.4]$ | 150 | 1623 | 990 | 1390 |
| Ga metal | [29.5 → 0] | 250 | 148 | 30 | 2204 |

Table 2. Summary of the target parameters. Energy values were recalculated with respect to corrected RbCl density assumptions

| | Maximum Beam Current | Target | Capsule | Energy window (MeV) [recalculated] |
|--------------------|-------------------------|--------|---------|--|
| Dummy stack | 250 μΑ | Nb | NONE | 93.5 – 71.4 |
| | | Zn | NONE | 66.5 – 43.1 |
| | | Al | NONE | 35.5 – 7.5 |
| Prototype Stack | 100 μΑ | RbCl | 316 SS | 92.4 – 72.7 [92.4 – 77.7] |
| | | RbCI | 316 SS | 65.1 – 45.1 [70.5 – 56.7] |
| | | Ga | Nb | 33.4 – 11.4 [47.4 – 33.4] |

From the results of the CFD analysis, the thermal conductivity-temperature curve of molten RbCl was chosen to be the "Nagasaka" data curve, where

$$k[W(mK)^{-1}](T) = 0.249 - 1.1E - 4 (T - T_m)$$
, and $T_m = 990K < T < T_b = 1390^{\circ}C$

These "Nagasaka" thermal conductivity data are lower than other reported data, and the choice leads to conservative estimates of the maximum beam current allowable for the RbCl salt target.

Table 3. Measured radionuclide yields of the three "Prototype" targets.

| Target | Isotope | Half-life (d) | Ex | yield) | |
|----------------|---------|------------------|-------|------------|-------|
| | | | Run#1 | Run#2 | Run#3 |
| | | | | | |
| High | Sr-82 | 25.5 | 72 | 76 | 58 |
| Energy | Sr-85 | 64.9 | 20 | 18 | 8 |
| RbCl | Rb-83 | 86.2 | - | 100 | - |
| | Rb-84 | 32.8 | - | 185 | - |
| | Rb-86 | 18.7 | - | 110 | - |
| | Br-77 | 2.375 | - | 300 | - |
| | Se-75 | 119.64 | - | 1.1 | - |
| | As-74 | 17.77 | - | 1.2 | • |
| | P-32 | 14.28 | - | 85 | 58 |
| | P-33 | 25.34 | - | - | - |
| Meduim | Sr-82 | 25.5 | 111 | 128 | 140 |
| Energy RbCl | Sr-85 | 64.9 | 22 | 28 | 26 |
| | Rb-83 | 86.2 | - | 125 | - |
| | Rb-84 | 32.8 | - | 203 | - |
| | Rb-86 | 18.7 | - | 115 | - |
| | P-32 | 14.28 | - | 26 | 19 |
| | P-33 | 25.34 | - | - | - |
| Gallium | Ge-68 | 270.82 | - | 8.2 | 8.5 |

Only the "Dummy" stack received $250\mu A$ of beam for a short time; it was irradiated for 4d and received a total charge amount of 9000 μAh during this period; "Prototype" stack #1, 2 d, 3142 μAh ; "Prototype" stack #2, 4 d, 9600 μAh ; "Prototype" stack #3, 43 h, 3714 μAh .

The three targets of the "Prototype" stacks (two RbCl disks and one Ga target) were chemically processed according to procedures published earlier [4]. Radioactive assay results are shown in Tab. 3.

Conclusion

Since one niobium encapsulated gallium target developed a blister after the extended irradiation of 4 days, a further evaluation of the gallium targets is required. Beside this gallium target, no other target showed any sign of thermal failure. Considering the uncertainties involved, the production yields obtained for targets irradiated in the same energy slot are consistent for all three "Prototype" stacks.

A careful analysis of the temperature profile in the RbCl targets shows that energy shifts occur in the RbCl and Ga targets. Energy shifts are a result of density variations in the RbCl disk under bombardment. Thickness adjustments of targets in the prototype stack are required to ensure maximum production yields of ⁸²Sr and ⁶⁸Ge in the design energy windows.

The ⁶⁸Ge yields obtained are still consistently lower than the predicted [5] yield value, which requires further investigation. After recalculation of the energy windows for the RbCl and Ga targets, the measured ⁸²Sr production yields compare rather well with values predicted on the basis of evaluated experimental excitation function data [5,6].

Acknowledgements

The construction project, the targetry R&D, and the isotope production efforts are supported by the U. S. Department of Energy, Office of Isotopes for Medicine and Science.

References

- 1. D. R. Phillips, R. C. Heaton, F. M. Nortier, M. E. Fassbender, E. J. Peterson and F. O. Valdez, The New 100 MeV Isotope Production Facility at LANL. *Transactions of The American Nuclear Society and The European Nuclear Society*, **TANSAO 89 1–920** (2003) p.769.
- 2. F.M. Nortier, M.E. Fassbender, R.C. Heaton, J.W. Lenz, E.J. Peterson and D.R. Phillips, High Current Targetry for the 100 MeV Isotope Production Facility, *Transactions of The American Nuclear Society and The European Nuclear Society*, **TANSAO 89 1–920** (2003) p.770.
- 3. J. Lenz and M. Nortier, CFD Flow and Thermal Analysis of LANL IPF Targets (These Proceedings).
- 4. M.Faßbender, F.M. Nortier, D.R. Phillips, V.T. Hamilton, R.C. Heaton, D.J. Jamriska, J.J. Kitten, L.R. Pitt, L.L.Salazar, F.O. Valdez, E.J. Peterson, Some nuclear chemical aspects of medical generator nuclide production at the Los Alamos hot cell facility, Radiochim. Acta 92, 237 (2004).
- 5. S.M.Qaim, F.T.Tarkanyi, P.Oblozinsky, K.Gul, A.Hermanne, M.G.Mustafa, F.M.Nortier, B.Scholten, Y.Shubin, S.Takacs and Y.Zhuang, *Charged-Particle Cross Section Database for Medical Radioisotope Production*, **IAEA-TECDOC-1211** (May 2001).
- 6. E.Z.Buthelezi, The Determination of Excitation Functions for ^{nat}Rb + p up to 100 MeV with an Emphasis on the Production of ⁸²Sr. MSc thesis, University of Western Cape, South Africa (January 2000, Unpublished).